Materials Physics Department

Physical and Engineering Sciences Center

Importance of Material Interfaces

Meeting the nation's critical challenges—whether in weapons systems, homeland security, or alternative energy—requires innovative, high-performance materials. Sandia's Materials Physics Department conducts the basic science that makes possible future innovations in materials engineering.

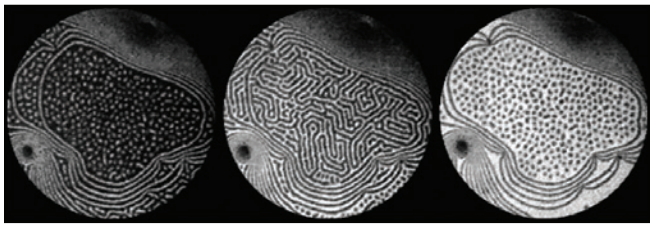
Because a material's performance is strongly influenced by its interfaces—both external surfaces and internal interfaces such as grain boundaries—we are pursuing an intensive research program to determine the atomistic processes responsible for macroscopic interface structure. In particular, we study the structure and motion of interfacial dislocations, impurities, surface steps, and phase boundaries.

By tightly integrating theory with experiment, we are uncovering the fundamental mechanisms that control the formation and stability of interfaces and that dictate how interfaces interact with their surrounding environment. In theoretical studies, we use first-principles electronic structure calculations to understand the mechanisms of atomic motion. Experimentally, we use advanced electron microscopies to characterize interfacial structure. With this combination we have achieved new understandings of the basic processes that control interfacial behaviors such as surface alloying, thin-film de-wetting, and grain boundary defaceting.

Surface Dynamics

As one of our overarching research directions, we are quantifying the fundamental atomic processes that govern the dynamics of surface structure and morphology. We use state-of-the-art, real-time microscopy—such as low-energy electron microscopy (LEEM) and scanning tunneling microscopy (STM)—to measure the time evolution of carefully defined nanoscale surface structures. We then compose thermodynamic equations of motion that describe this evolution, and relate the parameters of our models to atomic processes using statistical mechanics and density functional theory (DFT).

This approach has afforded new and often surprising insights into the mechanisms that underlie surface alloying, metal oxidation, thin-film de-wetting, two-dimensional self-assembly, surface motion, thermal surface smoothing, and misfit dislocation.



Pattern self-assembly: Through real-time microscopy, we have shown that nanoscale domain patterns on certain surfaces are caused by competition between short-ranged atomic attractions and longer-ranged repulsions. The repulsions arise from different sources, depending on the substances involved. Here, LEEM images show the self-assembled patterns that occur after depositing increasing amounts of Pb on Cu(111). The dark phase is a random Pb-Cu surface alloy, the bright phase is a Pb overlayer. (Temperature is 675K, field of view is 4 mm.)

Building upon this understanding, we now seek to gain the quantitative knowledge that scientists will need in order to engineer surface properties with predictable precision.

Metallic Interfaces and Dislocations

Interfacial defects are key to understanding how the overall configuration of an interface is determined by the atomistic details of its structure and composition. Just as bulk crystal behavior is dominated by the properties of lattice defects, many interfacial processes are controlled by point defects (such as vacancies and impurities) and extended defects (such as steps, dislocations, and junctions). Accordingly, we seek to explain how the lattice and chemical misfits that arise at interfaces are accommodated, and the implications of these relaxations for interfacial structure and behavior.

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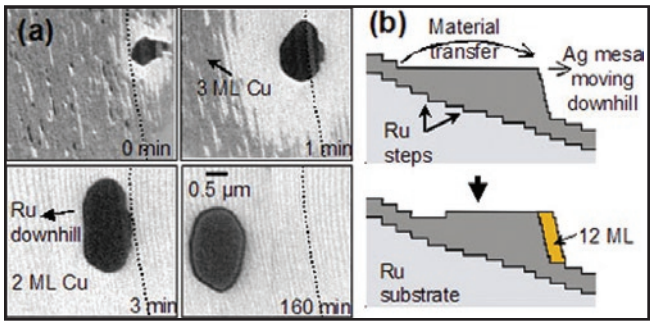
Again, we combine detailed experimental observations—using high-resolution TEM, STM, and atom probe tomography—with theory and computation, drawing upon continuum elasticity, interfacial crystallography, atomistic simulations, and first-principles simulations.

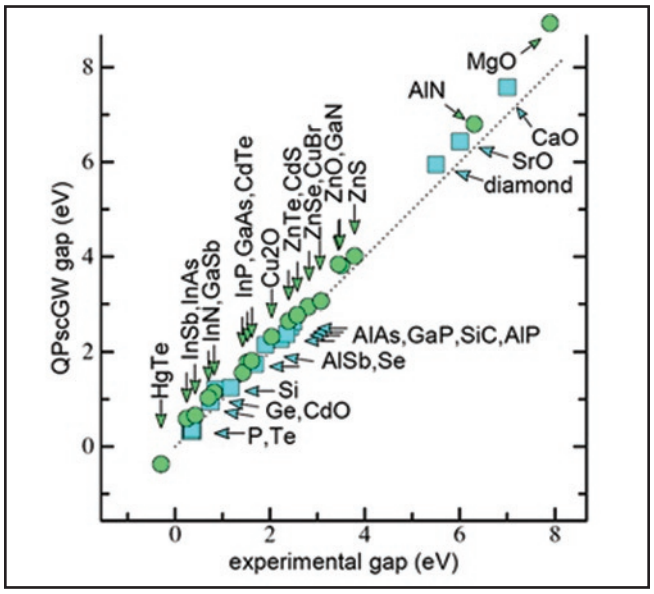


Film de-wetting: Thin films often fail to wet their substrates uniformly because three-dimensional islands can form, exposing bare substrate. Our group elucidated the energetics behind this puzzling island formation. A "downhill migration" process using steps in the substrate allows islands to form without having to nucleate a new layer. The LEEM images (a) and schematics (b) show Cu and Ag island movement—and attendant growth—on a Ru substrate.

This work has given us a fuller, dislocation-based understanding of interfacial structure, which has, in turn, led to new insights. For instance, we have shown how the competition between interfacial energy and strain energy can strongly influence boundary structure, inducing a structural transition in nanoscale boundaries. We have explained the atomic mechanisms that control the grain boundary defaceting transition, demonstrating that interfacial stress is not a critical factor in this case. We have learned how strains at grain boundaries can be accommodated by step-like defects. And we have discovered how complex, 3D interfacial reconstructions into non-bulk-like arrangements can be explained in terms of interfacial dislocation structures.

Electronic Transport in Nanostructures

In addition to exploring interfacial phenomena, we are also performing theoretical and experimental work to gain a fundamental understanding of electronic transport in nanostructures such as carbon nanotubes, nanowires, and single molecules. These studies combine synthesis, assembly, and electrical and optical characterization, with ab initio calculations and continuum modeling, to identify the mechanisms that control electronic transport, electrical contacts, and photocurrent generation. For example, we have recently shown that electronic transport in GaN nanowires is dominated by space-charge-limited current, a transport regime that was unexpected. In addition, we demonstrated that electrical contacts to carbon nanotubes and nanowires behave much differently than contacts to bulk systems.



Calculating electronic structure from first principles: Our quasiparticle self-consistent GW (QPscGW) approximation generates a one-particle potential capable of calculating reliable quasiparticle excitation energies for most of the periodic table. This ab initio approach is much more accurate than existing approaches for solid-state physics (LDA, RPA, GW). The graph compares the fundamental gaps calculated using our QPscGW theory to experimental results for sp compounds.

Emerging Research Directions: Thermoelectrics and Heteroepitaxial CVD Diamond

We are seeking to improve fundamental understanding of formation processes for thermoelectric alloys and the mechanisms by which their thermoelectric properties are enhanced. In another effort, we are developing heteroepitaxial, chemical-vapor-deposited diamond as a sensor for fission-spectrum neutrons (with potential applications in homeland security and nuclear nonproliferation).

Learn more at: http://public.ca.sandia.gov/8700

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